Carbon Dioxide Capture from Flue Gas Using Dry, Regenerable Sorbents

Quarterly Technical Progress Report

Reporting Period:
January 1, 2006 to March 31, 2006

by
David A. Green
Thomas O. Nelson
Brian S. Turk
Paul D. Box
Raghubir P. Gupta

DOE Cooperative Agreement No. DE-FC26-00NT40923

Submitted by:

Research Triangle Institute
Post Office Box 12194
Research Triangle Park, NC 27709-2194

April 2006

Disclaimer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United State Government or any agency thereof. The views and opinions of authors expressed therein do not necessarily state or reflect those of the United States Government or any agency thereof.

Abstract

This report describes research conducted between January 1, 2006, and March 31, 2006, on the use of dry regenerable sorbents for removal of carbon dioxide (CO_2) from coal combustion flue gas. An integrated system composed of a downflow co-current contact absorber and two hollow screw conveyors (regenerator and cooler) was assembled, instrumented, debugged, and calibrated. A new batch of supported sorbent containing 15% sodium carbonate was prepared and subjected to surface area and compact bulk density determination.

Table of Contents

Section **Page**

List of Figures

Figure **Page**

List of Tables

Table **Page**

1.0 Executive Summary

The objective of this project is to develop a simple and inexpensive process to separate carbon dioxide (CO_2) as an essentially pure stream from a fossil fuel combustion system using a regenerable sorbent. The sorbents being investigated in this project are alkali carbonates, particularly sodium carbonate (Na_2CO_3) which is converted to bicarbonate or to an intermediate salt through reaction with CO_2 and water vapor. The sorbent is regenerated to carbonate when heated, producing a nearly pure CO_2 stream after condensation of water vapor.

An integrated bench-scale system was assembled, instrumented, debugged and tested. The system is composed of two distinct process loops; the downflow co-current contact absorber for contacting flue gas with carbonate-based sorbent and two vertical hollow screw conveyors, to regenerate (heat) and cool the sorbent. The first screw thermally regenerates the sorbent, with heat supplied by condensing steam in the inside of the shaft and hollow jacket of the screw. The CO_2 and H_2O released during regeneration are disengaged from the sorbent at the top of the first screw. Condensation of the H_2O results in an essentially pure stream of CO_2 (which, in large-scale application, can be used for carbon sequestration or enhanced oil recovery). The second screw cools the sorbent to absorption temperature and returns the sorbent to the top of the downflow absorber. Sorbent cooling is achieved by indirect heat exchange using cooling water circulated inside the hollow jacket of the second screw.

An additional batch of supported carbonate-based sorbent was produced. This material was characterized and found to have consistent physical properties with earlier batches of sorbent.

2.0 Introduction

Fossil fuels used for power generation, transportation, and non-utility sectors are the primary sources of anthropogenic CO_2 emissions. Although there are many potential approaches to limiting these greenhouse gas (GHG) emissions, including increased energy efficiency and use of carbon-free fuels, it is clear that CO_2 capture and sequestration will play an important role in mitigating the progress of global warming. In the near future, CO_2 capture efforts will likely focus on large stationary sources, such as fossil-fueled power plants because these sources emit the largest quantities of CO_2 and will offer the benefits of economy of scale. It is for this reason that the United States Department of Energy's (DOE's) Carbon Sequestration Program, administered by the Office of Fossil Energy and managed by the National Energy Technology Laboratory (NETL), conducts and funds research to develop CO_2 capture and sequestration technologies.

The focus of this project is the development of a simple and inexpensive process to remove CO_2 from the flue gas of existing power plants (particularly coal-fired plants) using a dry, regenerable sorbent. This capture technology is based on the reversible reactions between CO_2 and sodium carbonate. Using a cyclic thermal-swing process, an essentially pure CO_2 stream can be removed from flue gas for subsequent sequestration or reuse. Capture of CO_2 from low-temperature flue gas using Na_2CO_3-based sorbents results in the reversible formation of sodium bicarbonate ($NaHCO_3$) and/or Wegscheider's salt ($Na_2CO_3 \cdot 3NaHCO_3$), as shown in Reactions 1 and 2:

Reaction 1

$$Na_2CO_3(s) + CO_2(g) + H_2O(g) \leftrightarrow 2NaHCO_3(s) \qquad \left[\Delta H_r^o = -32.4 \text{ kcal/mol } CO_2\right]$$

Reaction 2

$$Na_2CO_3(s) + 0.6\ CO_2(g) + 0.6\ H_2O(g) \leftrightarrow 0.4\left[Na_2CO_3 \bullet 3NaHCO_3(s)\right] \qquad \left[\Delta H_r^o = -32.5 \text{ kcal/mol } CO_2\right]$$

Both forward reactions (CO_2 absorptions) are exothermic. The equivalent reverse reactions (sorbent regeneration) are endothermic and produce equal molar quantities of CO_2 and H_2O. Condensation of H_2O from the regeneration product results in a pure CO_2 stream that is suitable for sequestration or reuse.

This report describes activities conducted between January 1, 2006, and March 31, 2006, by RTI International (RTI). Activities conducted this quarter include assembly, instrumentation, debugging and calibrating an integrated downflow absorber/sorbent regenerator/sorbent cooler system.

3.0 Experimental

3.1 Assembly of Integrated System: Downflow Absorber and Screw Conveyors

The screw conveyors were received in mid-January and installed into the frame built in the previous quarter. Screw conveyor design specifications were described in the previous quarterly report (Green, et al., 2006). The entire unit was assembled with all supporting brackets, steam and condensate piping and associated equipment, instrumentation, electronics, and electricity supply. Sorbent conveying rate was determined as a function of screw rotation rate.

The screws that were originally supplied by the vendor were fitted with ¼ HP motors and could only produce a maximum eight revolutions per minute. The maximum flow rate attained was only about 45 lb/hr, much lower than requested. The specifications called for a maximum solid flow of 250 lb/hr. It was determined that higher speeds would be required to produce the desired solid flow rate. New, 2 HP motors were installed in addition to a gear reducer capable of delivering a maximum speed of 45 RPM. Larger variable frequency drives (VFDs) were also installed to accommodate the increased electrical load required by the new drives (Note: Additional testing to accurately determine the sorbent conveying rate versus screw rotation speed were conducted in April 2006 and thus will be reported in the next quarterly report).

During the initial flow tests using the new motors and gear reducers, it was determined that the shaft seals originally installed were inadequate for the application. These seals have been replaced by more robust, 'multi-protection' Martin Sprocket Super Pack ™ shaft seals. Preliminary testing indicates that these seals are more than adequate for application on the integrated unit.

Preliminary testing was conducted with simulated flue gas and precalcined carbonate-based sorbent (RTI supported sorbent). The integrated system, as presently configured is shown in Figure 1. An updated construction drawing of the screw conveyor system is shown in Figure 2. An updated parts list for the screw conveyor system is given in Table 1.

Figure 1. Integrated Carbon Dioxide Capture System: Downflow Contactor and Screw Conveyors.

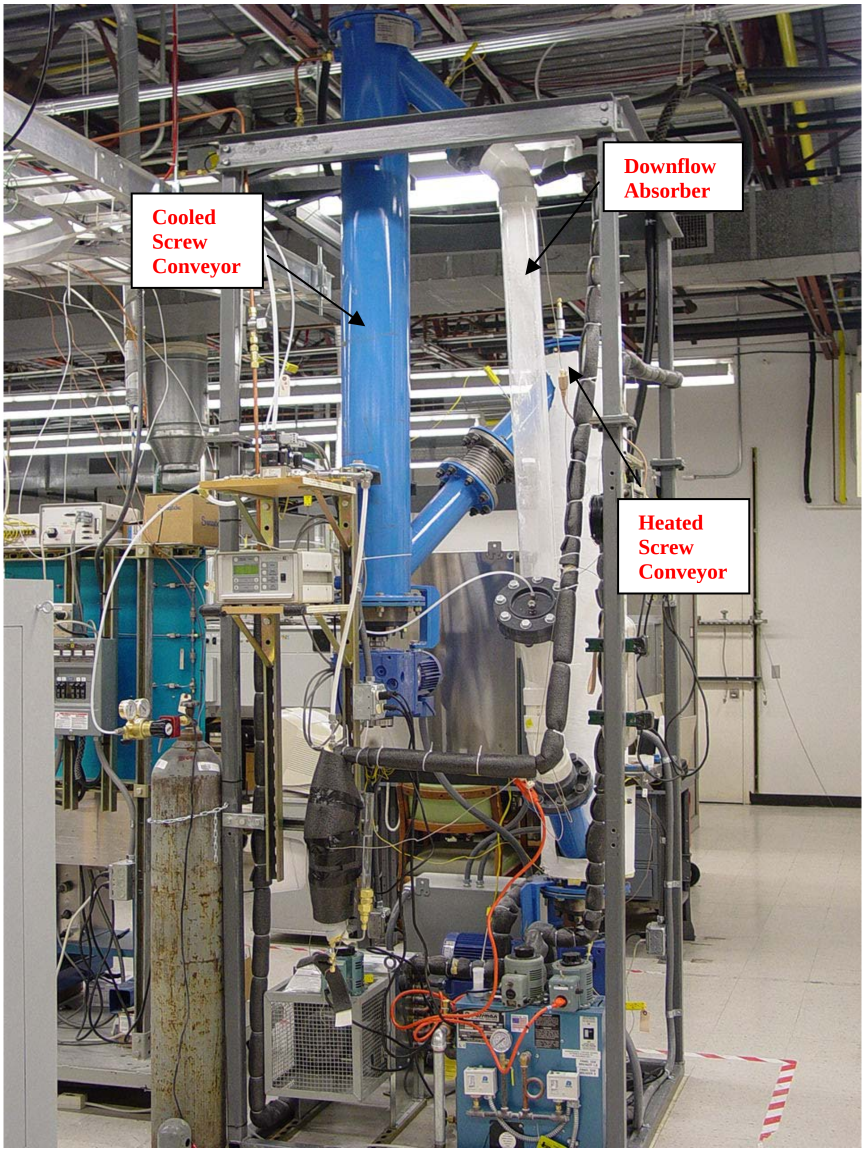


Figure 2. Updated Construction Drawing of Screw Conveyors.

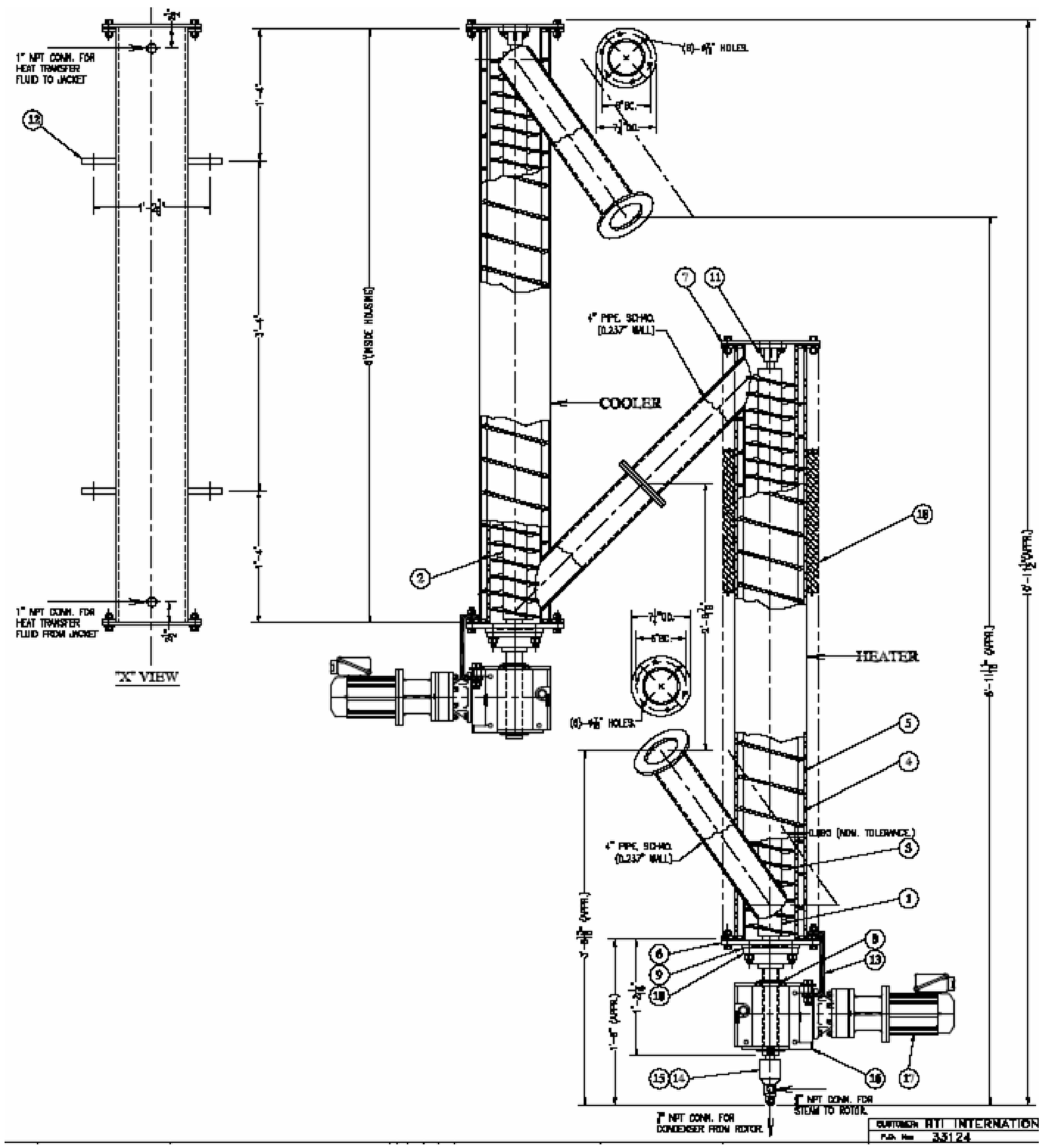


Table 1. Updated Parts List and Specifications for Integrated System.

TLV-20767-C	1	1	Heater Rotor, Hollow Drive Shaft, 5 7/8’ NOM, OD, w/ 2’ Pitch & 2’ Stem Pipe	CS
TLV-20768-C	2	1	Cooler Rotor, Sold Drive Shaft, 5 7/8’ NOM, OD w/ 2’ Pitch & 2’ Stem Pipe	CS
TLV-2069-C	3	2	Tubular Housing, 6’ Pip Sch 40 (0.28’ wall)	CS
TLV-2069-C	4	LOT	Helicoidal Support	CS
TLV-2069-C	5	2	Housing Jacket, 8’ Pipe Sch 20 (0.25’ Wall)	CS
TLV-15939-A	6	2	End Plate, (Drive End)	CS
TLV-15940-A	7	2	End Plate, (Tail End)	CS
	8	2	External Retaining Ring, # 5100-206	STEEL
	9	2	Super Pack Seal, ‘Martin’ Part # MSP4 (2’ Bore)	–
	10	2	Flange Ball Bearing, ‘Martin’ Part # TEB4BB (2’ Bore)	–
	11	2	Flange Bronze Bearing, ‘Martin’ Part # TEB2BR, (1’ Bore)	–
TLV-20769-C	12	8	Lifting/Holding Lug.	CS
TLV-15940-A	13	2	Gear Reducer Support	CS
	14	1	Rotary Joint	–
TLV-159641-A	15	1	Rotary Joint Stop, (Not Shown)	CS
	16	2	Shaft Mounted Gear Reducer, Ratio 60:1	–
	17	2	2 hp 2 ph MOTOR	–
	18	1	1 1/3’ Thk. Fiberglass Pipe (Insulation w/ Self-Sealing Jacket)	–

3.2 Production of Additional Sorbent

An additional 300 pounds of sodium bicarbonate were received from Solvay Chemicals for use in shakedown and preliminary testing of the bench-scale screw integrated system. An additional 80 pounds of RTI’s “CO_2 Capture Sorbent” were produced by Süd-Chemie, Inc for testing in the integrated system. This batch of sorbent was subjected to BET surface area and compact bulk density determinations.

4.0 Results and Discussion

4.1 Screw Conveyor Shakedown and Calibration

One of the first shakedown procedures conducted on the integrated unit was the calibration of the original 0.25 HP motors and speed controllers. The correlation between motor frequency and rotational speed of the shafts was found to be linear. These data are shown in Figure 3. The sorbent flow rate with these motors is shown in Figure 4. Sorbent flow rate appears to be linear with respect to screw conveyor RPM. Sorbent flow rate was determined by ducting the sorbent discharged by the cooling screw to a bucket resting on a digital electronic balance and determining the mass of solids collected over a given time interval. The maximum achievable sorbent flow rate was much less than the desired, manufacturer-specified flow rate. The motors, gearboxes and speed controls were then replaced with higher capacity equipment to solve this issue.

Figure 3. Speed Control Correlation.

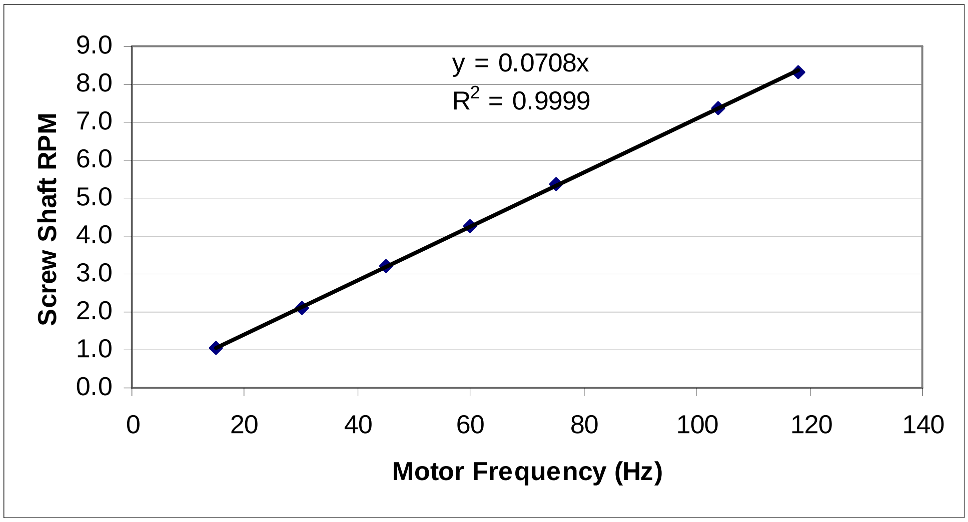
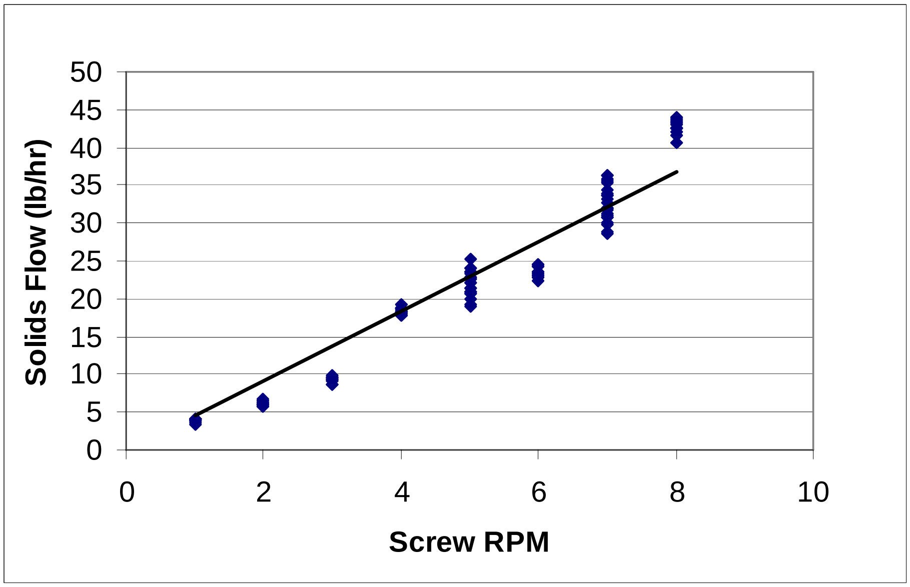


Figure 4. Solids Flow Rate: 0.25 HP Motors.



The solids flow rate with the 2 HP motors, new gearboxes, and speed controls installed is being determined at the time of this writing. Based on prelimary data, RTI was able to flow approximately 150 pounds per hour of sorbent at a screw rotational speed of 45 RPM. This result is much closer to the flow rates desired in the integrated system. RTI continues to shakedown the integrated system and solve problems that arise during this shakedown.

4.2 Characterization of New Batch of Sorbent

SCI produced 80 pounds of supported sorbent containing 15% Na_2CO_3 on ceramic support. This material was given the name “SCI-022806-1”. A comparison of the properties of this material with the previous batch is shown in Table 2. The data show the material to be very similar to the previous SCI-produced batches and thus lends credit to the reproducibility of SCI’s manufacturing procedure.

Table 2. Comparison of Supported Sorbent Properties

Sorbent	SCI-012705-1	SCI-090905-1	SCI-022806-1
Na_2CO_3 Content, %	15	10	15
Surface Area, m^2/g	96.5	117	102
Bulk Density, g/cc	0.96	0.88	0.91

5.0 Other Project Activities

A presentation, covering progress to date and future project plans, was made at the Second Annual Capture and Transportation Working Group Workshop held on March 22-23, 2006 in Palo Alto, CA. An abstract for a paper to be presented at the Fifth Annual Conference of Carbon Capture and Sequestration in Alexandria, VA in May 2006 was submitted and accepted. An abstract for a paper to be presented at the Pittsburgh Coal Conference in September, 2006 was submitted.

6.0 Conclusions

Circulation of carbonate-based sorbents (RTI supported sorbent) in an integrated downflow absorber/sorbent regenerator/sorbent cooler system is feasible. CO_2 removal from simulated flue gas has been demonstrated using precalcined sorbent in the integrated system.

7.0 Future Work

Operation of the integrated system will be optimized and regeneration of sorbent in the heated hollow screw will be demonstrated, along with CO_2 recovery. A paper will be presented at the Fifth Annual Conference on Carbon Capture and Sequestration, in May 2006.

8.0 References

Green, D.A., Nelson, T.O., Turk, B.S., Box, P., Weber, A., and Gupta, R.P. 2006. *Carbon Dioxide Capture from Flue Gas Using Dry Regenerable Sorbents*. Quarterly Technical Progress Report, RTI International. January.